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Arthur D. Little, Inc.  
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Covering the Period  
July 15, 1961 to October 15, 1961

Research in Electrical Phenomena Associated with Aerosols

A S T I A

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QUARTERLY REPORT NO. 5

RESEARCH IN ELECTRICAL PHENOMENA ASSOCIATED WITH AEROSOLS

ABSTRACT

The fate of charge on a charged volatile particle was investigated. Charged droplets were held suspended in an electric field and allowed to evaporate. Charge and mass were measured as a function of time. Charged water drops evaporate until the electrical forces on the surface are equal to the surface tension forces at which time the droplets disrupt to eject a number of small highly charged droplets. The fraction of charge and mass lost with each ejection and the effect of maximum charging on evaporation rate of a drop are discussed.

Arthur D. Little, Inc.

## I. INTRODUCTION

A series of experiments have been performed to study the fate of the charge on a charged volatile particle. It has been observed in several instances that when a charged volatile particle evaporates the charge-to-mass ratio apparently increases. In some instances residues of extremely high mobility were observed which can only be accounted for by the presence of charge far in excess of expected maximum values. The object of our work during this quarter has been to closely examine the evaporation of a charged drop to determine what actually happens to the charge and what the important parameters in the evaporation process are.

## II. DESCRIPTION OF THE EXPERIMENT

The experimental technique we used was a variation of Millikan's oil drop experiment. A cloud of charged drops was injected into the electric field set up between two parallel horizontal plates. One of these drops was selected by eye and the voltage between the plates was adjusted so that the electric force on the particle just balanced gravity. At this condition the particle remained suspended in space and allowed the experimenter to observe the evaporation process for extended periods of time. During evaporation the mass decreased and the electric field between the plates was continually reduced to maintain the electrical force required to achieve the force balance.

The force balance is described by the relation:

$$mg = q E = q \frac{V}{d}$$

The following set of symbols and units are used henceforth:

$m$  = mass of drop in kg

$g$  = acceleration of gravity in  $\frac{\text{meters}}{\text{sec}^2}$

$q$  = charge on drop in coulombs

$V$  = potential between plates in volts

$d$  = distance between plate in meters

$\rho$  = density of drops in  $\frac{\text{kg}}{\text{meter}^3}$

$V_o$  = average potential at drop surface in volts

$E_o$  = average electric field at drop surface in  $\frac{\text{volts}}{\text{meter}}$

$\sigma$  = average charge density at surface of drop in coulombs per meter<sup>2</sup>

$e$  = charge on electron in coulombs

$\frac{\sigma}{e}$  = average number of electrons per meter squared on drop

$D$  = drop diameter in meters

Rearranging this expression we have

$$V = g d \frac{m}{q} \quad (1)$$

Since  $V$  and  $d$  can be directly measured and  $g$  is known we must obtain an independent value of  $m$  or  $q$  in order to solve for all variables in equation 1.

The experimental apparatus is diagrammed in Figure 1. The parallel plates were brass rectangles 36 cm by 40 cm separated by four glass insulators at the corners. The distance between the plates was

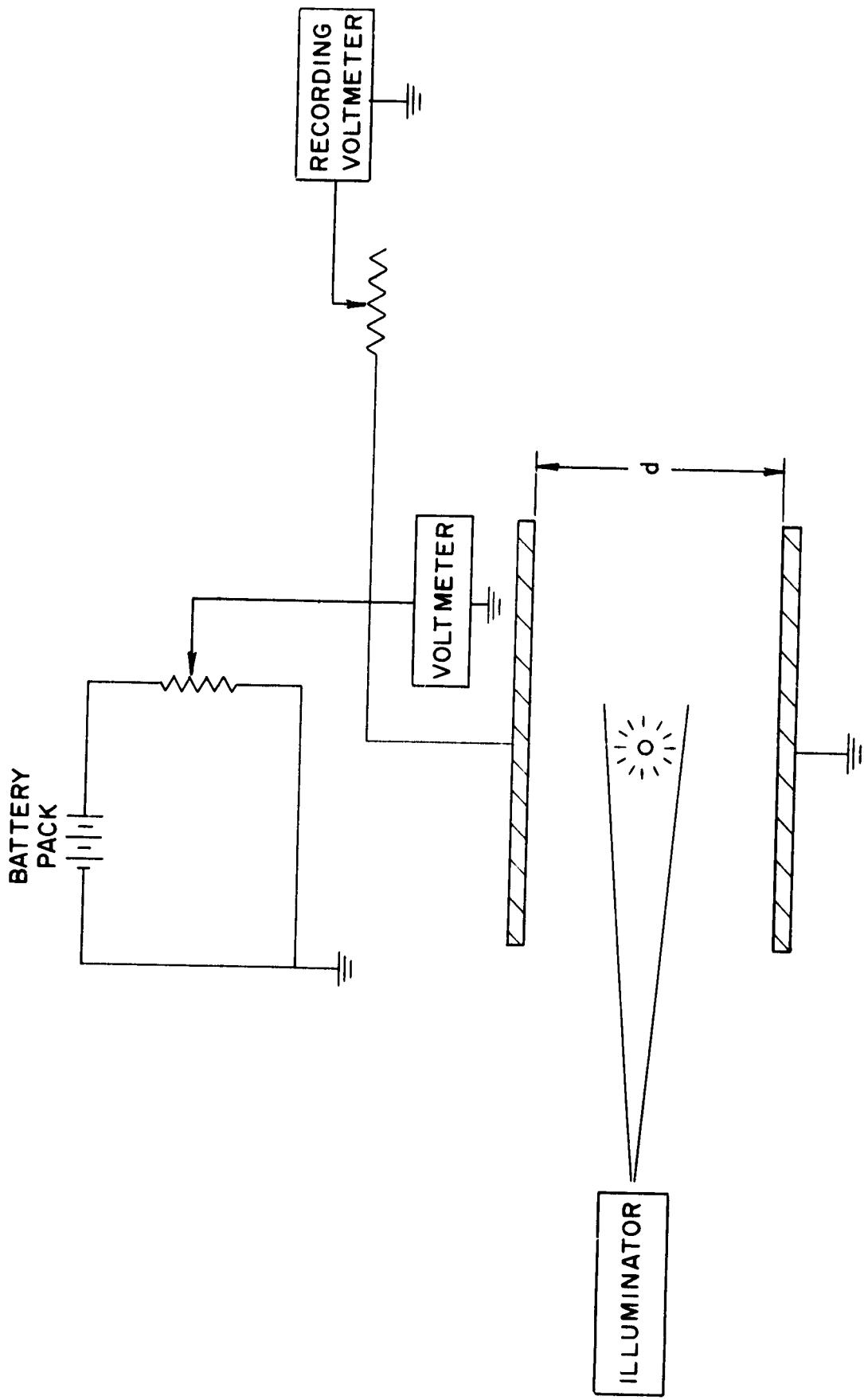


FIGURE 1  
SCHEMATIC OF TEST APPARATUS

11 centimeters. The plates were contained inside a box which was sealed except for a small hole through which the drops were injected.

Voltage was applied to one of the plates from a battery pack. It was controlled through a 5 megohm potentiometer. Normally, the initial voltage used was a few hundred volts.

A DC voltmeter indicated the voltage on the plates. An Esterline-Angus recorder was used to record plate voltage as a function of time. The meter had six ranges from 1.5 volts to 750 volts full scale. A one megohm variable resistor was used to protect the recorder meter movement when the low voltage ranges were used. This resistor was set to zero when the applied voltage was reduced to the full scale value.

Illumination was provided with a 300 watt slide projector. The charged cloud of drops was injected into the field between the parallel plates from a standard 30 cc hospital syringe. The hypodermic needle (23 gauge) was energized to about 5 KV with a 0 to 30 KV DC power supply. The drop sizes were measured in one of two ways. For larger drops (50 to  $130\mu$  diam.) the liquid was dyed and the drops were allowed to fall on a piece of filter paper after the required balancing voltage had been measured. The size of the spot on the filter paper was then measured under a microscope. The true drop size was read from a calibration curve of true drop size versus spot size. For smaller drops (20 to  $40\mu$  diam.) size was determined from time of fall measurements.

### III. RESULTS AND DISCUSSION

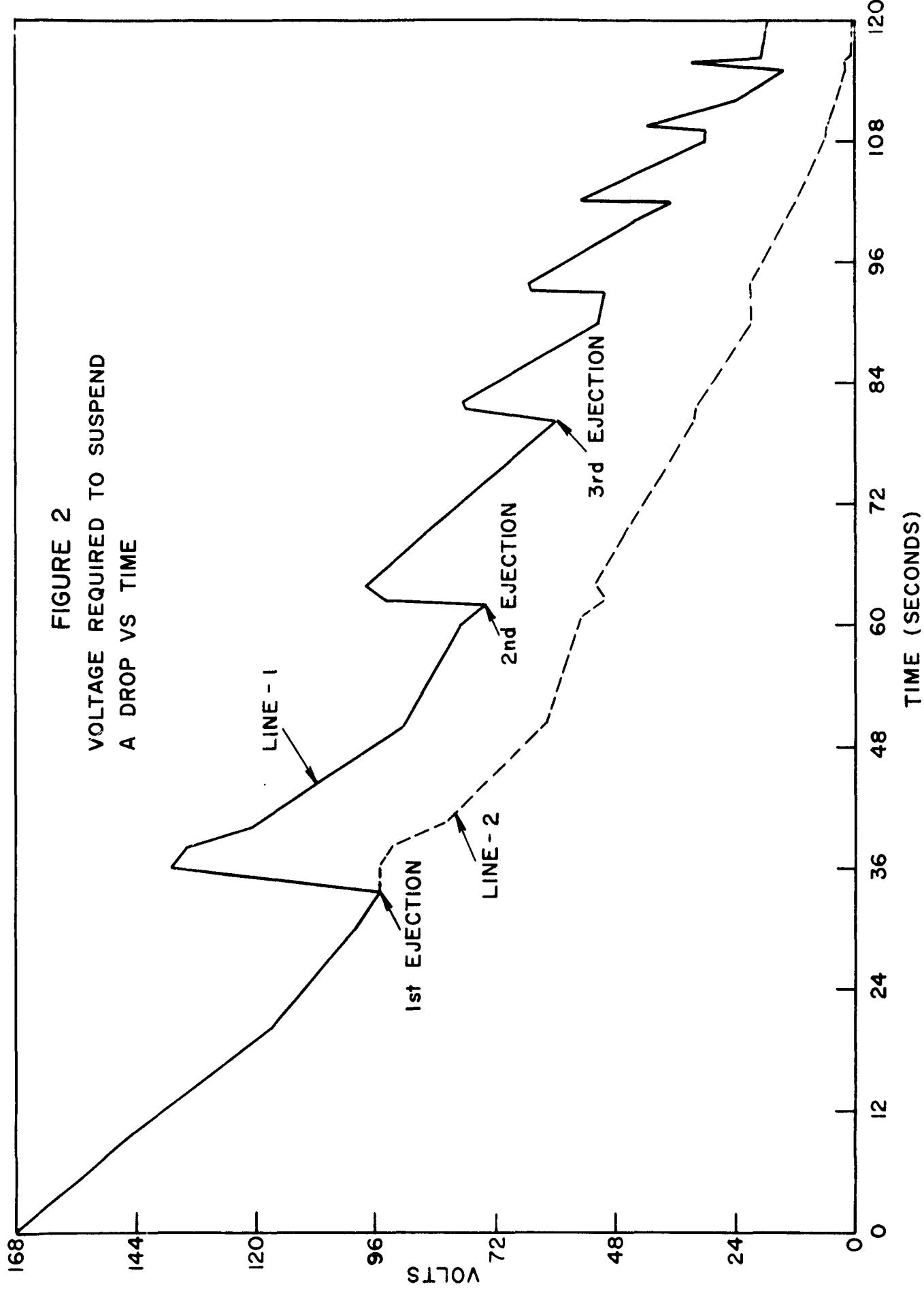
In a typical experiment a cloud of approximately 20 drops was injected into the electric field. These drops were of the order of 200 microns in diameter. One of these was selected by eye if the forces balanced and it was fairly stationary. The rest of the drops cleared

the field in a few seconds. As the drop evaporated the voltage was continuously decreased. After a period of time (usually less than 30 seconds) the drop suddenly fell from the field as if it had become very heavy. If the voltage was increased fast enough, the forces could again be balanced and the drop resuspended. The orderly evaporation would again proceed until the drop again fell. This fall-evaporate sequence has been observed to happen as many as eight times with a single drop. The heavy line in Figure 2 is a graph of the balancing voltage as it is controlled by the operator versus time showing the typical sawtooth pattern resulting from the fall-evaporate sequence. Note that the time interval between discharges grows shorter with each discharge. When the discharge number is plotted versus time on semilog coordinates an extremely regular performance is demonstrated as in Figure 3.

From equation 1 we see that a sudden fall implies a sudden decrease in  $q/m$ . What, in fact, was observed when the drop fell was the ejection from the parent drop of from one to around ten rather small drops. The  $q/m$  of these droplets was always substantially higher than the original drop. Their size was difficult to estimate, however, they probably ranged between one and fifteen microns. In general, the larger the initial drop the greater the number of ejected drops. A larger drop (roughly greater than  $100\mu$ ) ejected a small expanding cloud of drops directly upward. The smaller drops eject few droplets and they were ejected in any direction.

In the case of the smaller drops, a distinct recoil by the parent drop was observed. It is presumed that this also occurred in the case of the larger drops, however, as the recoil would have been downward and as the larger drops fall rapidly under gravity anyway it was difficult to know whether we were seeing gravity alone at work or gravity plus recoil.

FIGURE 2  
VOLTAGE REQUIRED TO SUSPEND  
A DROP VS TIME



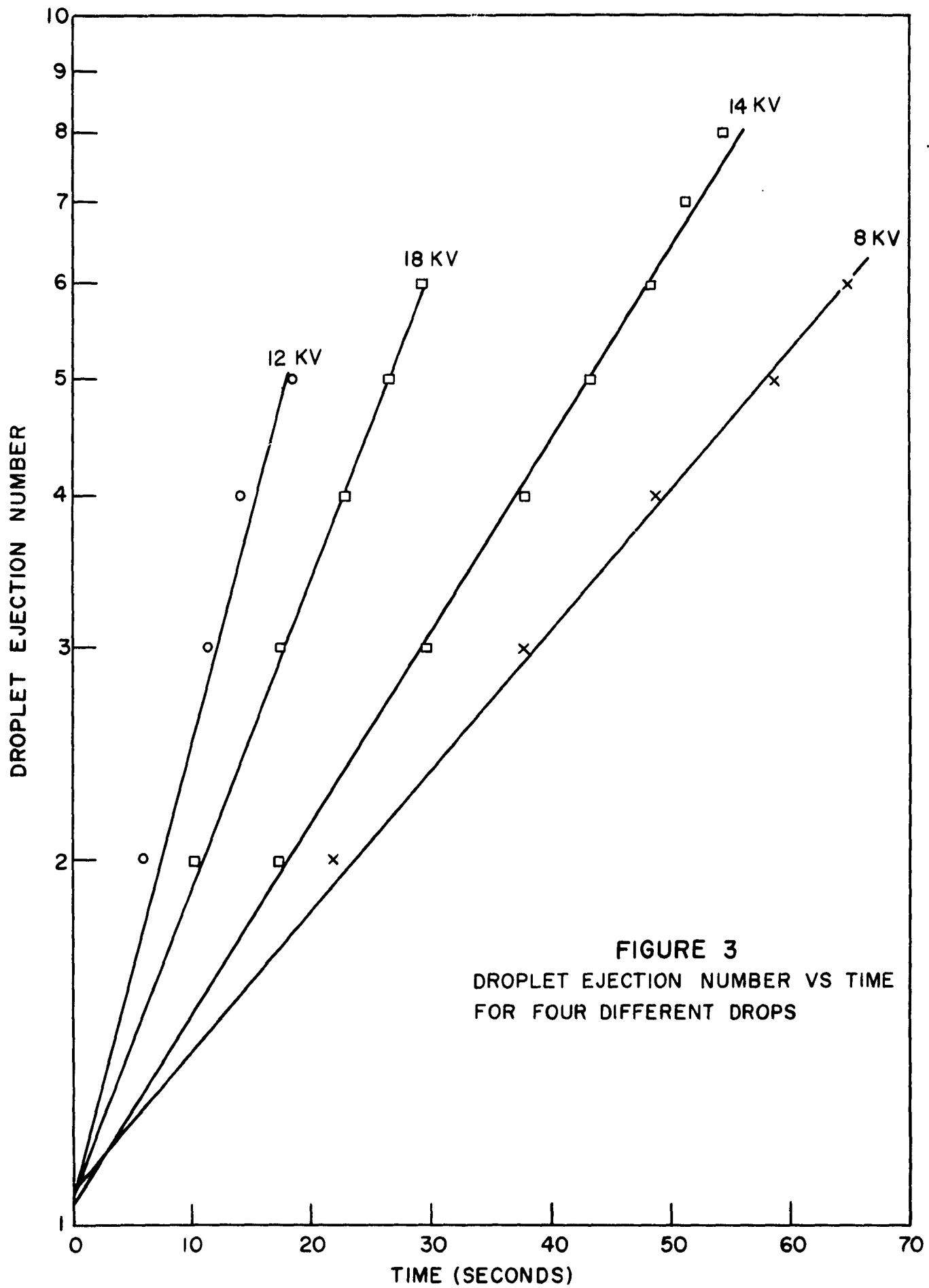


Figure 4 is a plot of balancing voltage immediately before drop ejection versus the balancing voltage immediately afterward. The graph clearly indicates a constant slope. An estimate of the upper limit of the number and mass of the ejected droplets and an estimate of the lower limit of the mass of the parent drop leads to the conclusion that it is extremely unlikely that the mass loss to the parent drop during droplet ejection is more than a few percent. Therefore, this constant voltage ratio implies that a constant fraction of the charge was lost with each event.

While at this point it is clear there are many interesting aspects of this question it was decided to restrict the investigation to the maximum charge density on the surface of a drop as a function of the drop diameter and drop fluid. Some conclusions can also be drawn on the effect of charge on evaporation rate.

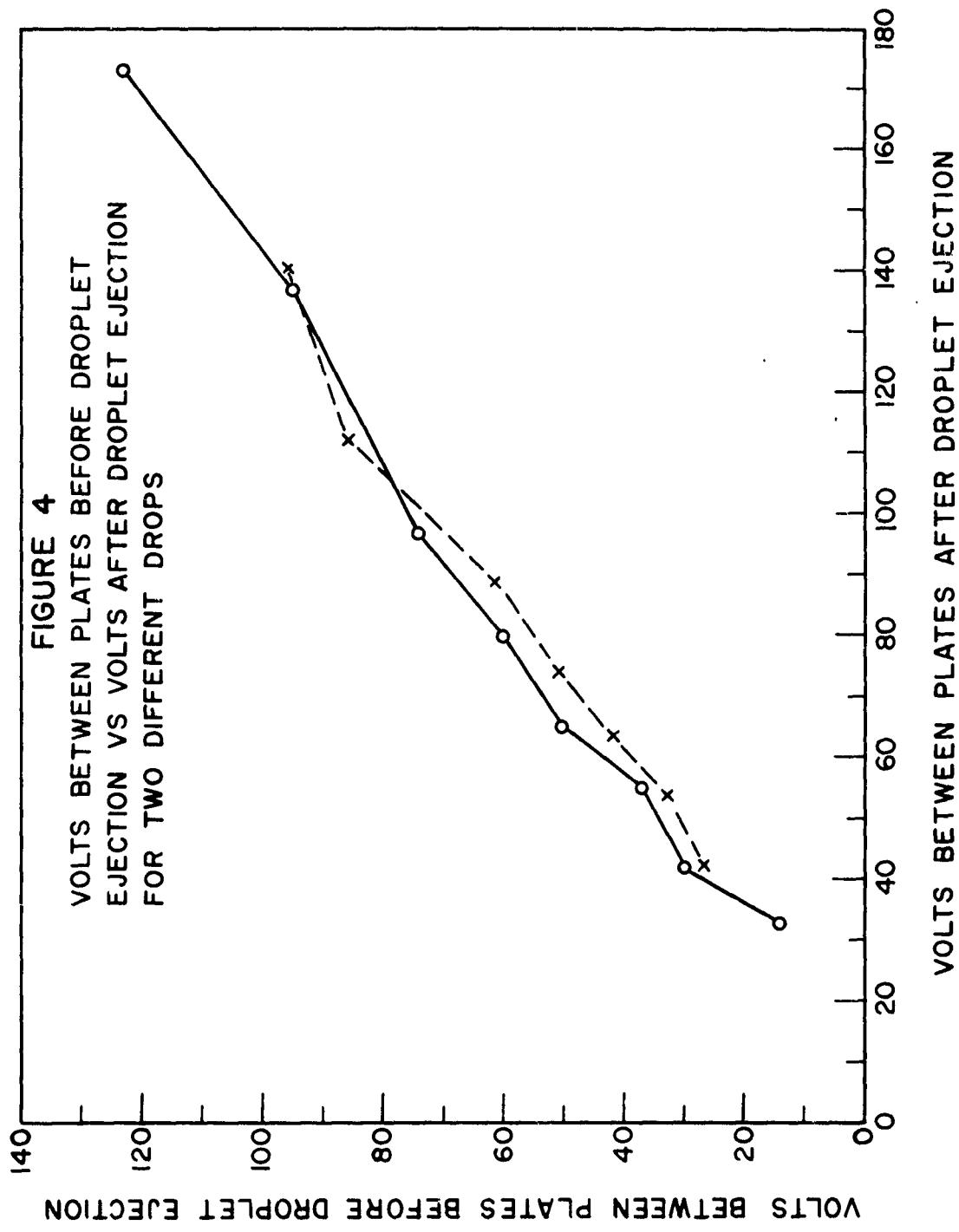
The effective surface tension ( $\gamma'$ ) of a uniformly charged spherical drop (see Quarterly Report No. 4) is

$$\gamma' = \gamma - \frac{\epsilon_0 E_0^2 D}{8}$$

(in c.g.s. units the expression is  $\gamma' = \gamma - \frac{\epsilon_0 E^2 D}{32\pi}$ ). When

$$\gamma = \frac{\epsilon_0 E^2 D}{8}$$

the electrical forces on the droplet surface would exactly equal the surface tension forces and droplet disruption could occur. Therefore, for any liquid drop of a given size the surface tension would control the maximum potential gradient one could achieve. Furthermore, the maximum potential gradient should vary or the inverse square root of the drop diameter.



In one series of experiments using colored water we allowed the highly charged drop from the hypodermic needle to evaporate until the droplet ejection took place and then as the parent drop fell no attempt was made to increase the voltage but rather it was allowed to fall on a piece of white filter paper so drop size could be determined. We recognize that in this way drop size was determined after an ejection but as stated before the mass loss was negligible with each event. From equation 1 it is easy to show that:

$$\frac{q}{\pi \epsilon_0 D^2} = \frac{\rho g d}{6 \epsilon_0} \frac{D}{V} = E_0 \quad (2)$$

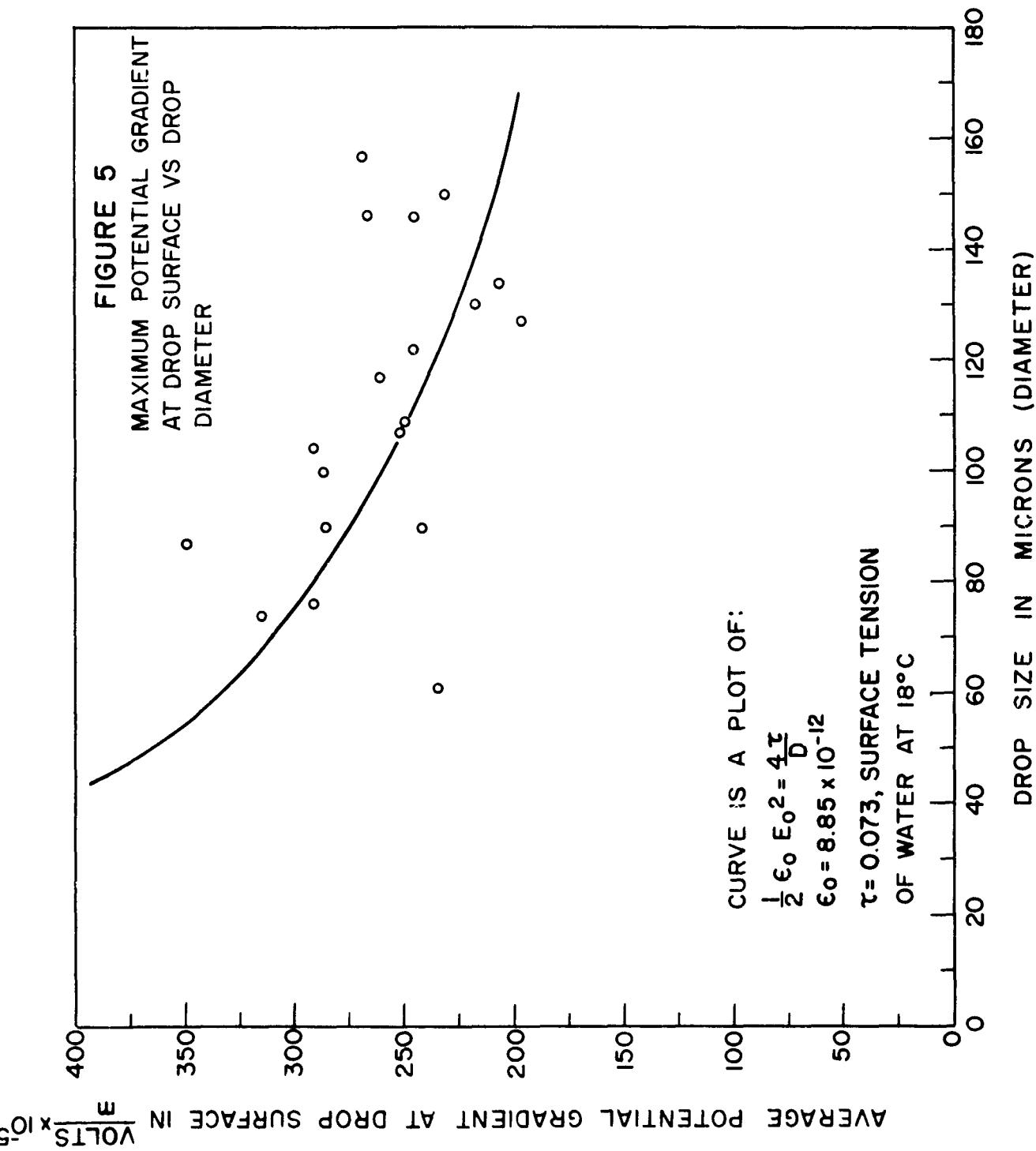
and

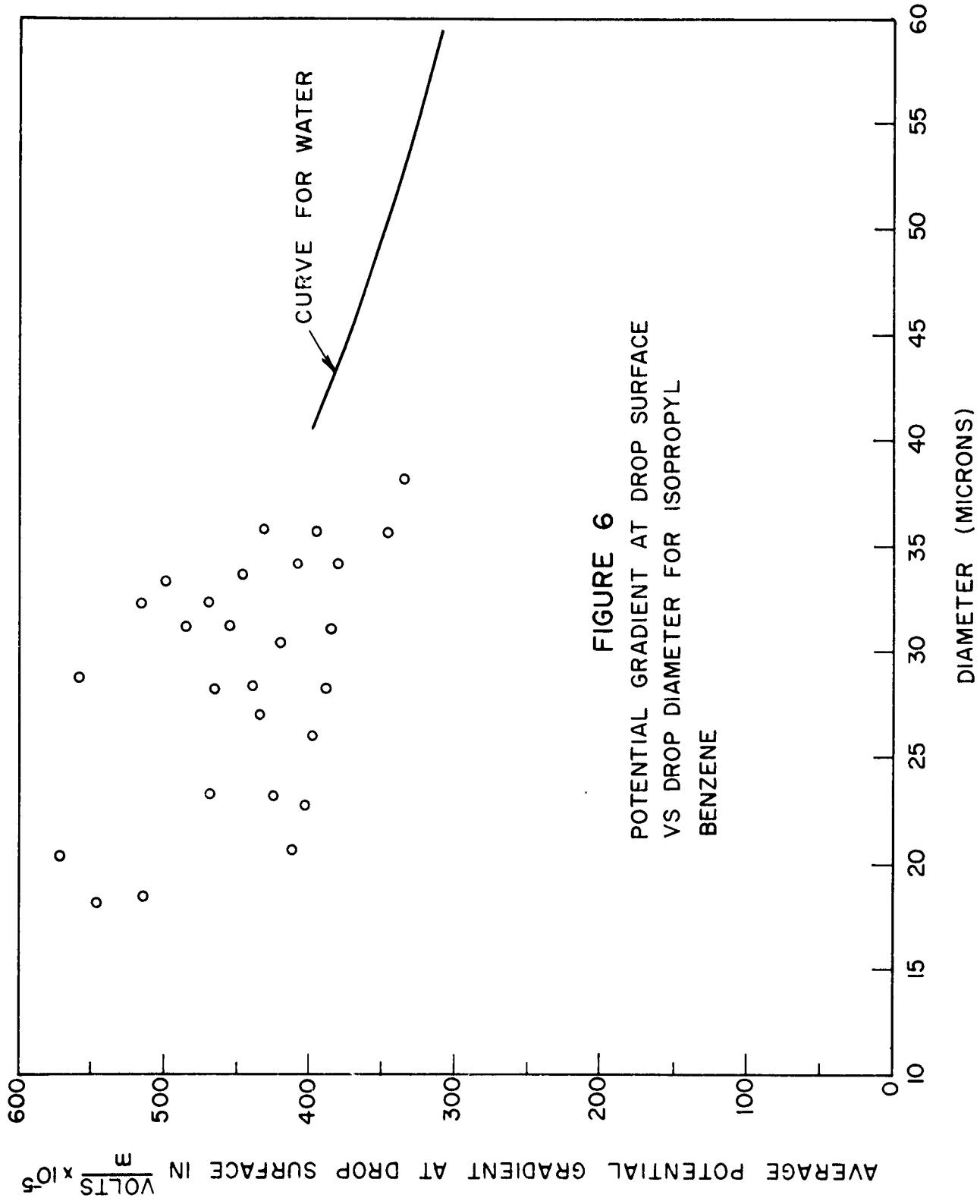
$$\frac{q}{\pi D^2} = \frac{\rho g d}{6} \frac{D}{V} = \sigma \quad (3)$$

If we assume that the charge on a drop does not distort its shape and that the charge is uniformly distributed on the surface then we can solve equation 2 for  $E_0$  and equation 3 for  $\sigma$ . Figure 5 is a plot of this data. The curve is the theoretical plot of  $1/2 \epsilon_0 E_0^2 = 4\tau/D$ . The correlation is extraordinary. Most of the scatter in the data is attributed to errors in the size measuring technique.

Later measurements were carried out using the time of fall technique. These experiments were limited by the equipment to a size range of from 15 to  $35\mu$ . Various organic fluids were used in place of water (aniline, isopropyl benzene and m-cresol). These were dissolved in acetone and injected in the usual manner. The acetone evaporated in a few seconds leaving a highly charged drop. Electric fields calculated for these drops do not represent maximum charge densities but rather the field at the time of measurement. Figure 6 is a plot of this kind of data for isopropyl benzene. The spread in the data is in part due to the fact that we are not always seeing maximum charging and also the measuring error. The important thing is that this data falls exactly on the curves of  $1/2 \epsilon_0 E_0^2 = 4\tau/D$  plotted in Figure 5 for water. However,

FIGURE 5  
MAXIMUM POTENTIAL GRADIENT  
AT DROP SURFACE VS DROP  
DIAMETER





the surface tension of isopropyl benzene is about 40% of that for water. Therefore, by comparison to water the benzene appears far too highly charged. This result was characteristic for all the organic fluids used in this size range.

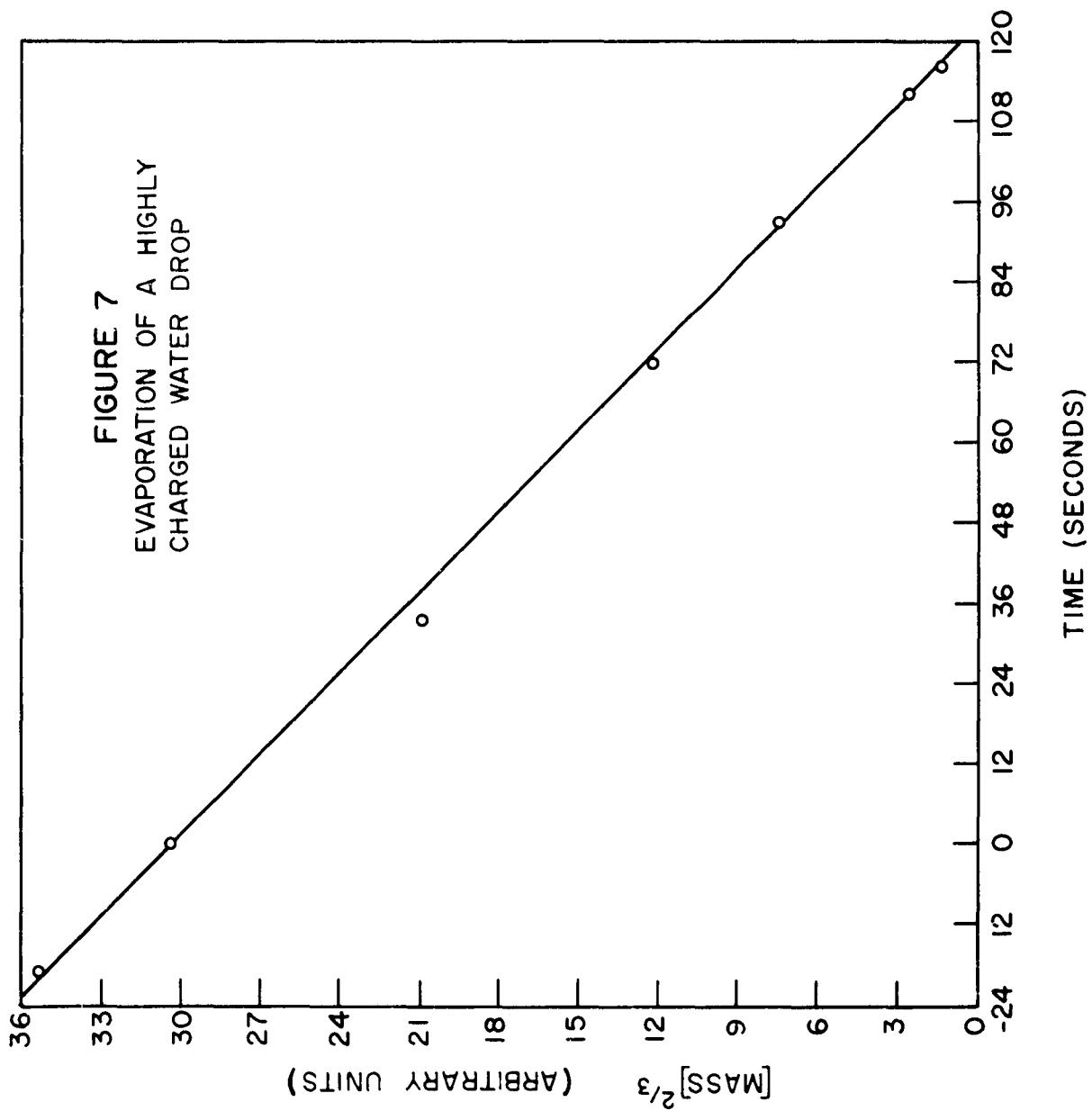
In Quarterly Report No. 4 the effect of charge on evaporation rate of drops in the size range of our study was shown theoretically to be negligible, therefore, we would expect to see no change in our experiments.

In order to evaluate the effect of charge on evaporation rate, we wished to obtain a curve of drop size versus time for a long period of time as the drop became increasingly charged. The sawtooth decay curve of voltage versus time of Figure 2 allows this. Between each droplet ejection the voltage curve is determined by the drop mass. When droplet ejection occurred the voltage curve was increased by the fraction of charge lost (since we have stated the mass loss was small). Therefore, if we multiply each successive segment of the sawtooth decay curve between droplet ejections by the total fraction of charge lost as measured by the product of all the preceding fractional voltage increases at each ejection, we will have a curve of drop mass versus time. The mass units, of course, are arbitrary. This was what was done to obtain line 2 of Figure 2.

The form of the evaporation curve for a spherical drop is:

$$m^{2/3} = A - Bt$$

In Figure 7 we have taken the line 2 of Figure 2 and plotted the  $2/3$  power of the mass (in arbitrary units) against time. The straight line fit is striking. Thus, we have not changed the shape of the evaporation curve. Significantly, the charge densities were near maximum where one would expect any effect to be most pronounced. Further-



more, the charge density charged during the evaporation. We have shown that  $\sigma \propto \frac{D}{V}$ . Also, the theory for maximum charging gives  $\sigma^2 \propto \frac{1}{D}$ . Therefore for maximum charging of a drop  $\sigma^3 \propto \frac{1}{V}$ . Since the ratio of voltage change for this curve is about eight, the change in charge density is a factor of 2. If there were any substantial effect of high charge densities on evaporation rate it should have been apparent with a factor of 2 change in charge density.

#### IV. CONCLUSIONS

The drop evaporates at constant charge until the electrical forces on the drop surface equal the surface tension forces. This relationship is expressed by:

$$\gamma' = \gamma - \frac{\epsilon_0 E^2 D}{8}$$

When the two terms on the right are equal, the droplet breaks up. One or more small charged droplets split off from the parent drop. The ejected droplets constitute a negligible fraction of the original drop mass, but a substantial fraction of the charge. After this event the parent drop continues to evaporate until the process repeats. Again the same fraction of charge is lost. The results for small drops of organic fluids are not quantitatively consistent with this explanation and further experimental work is required.

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